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ASSESSMENT OF GAS PRODUCTION AND PRESSURE CONDITIONS OF FIBRE REINFORCED CONCRETE CONTAINERS UNDER DISPOSAL DUE TO MICROBIAL DEGRADATION AND RADIOLYSIS OF CELLULOSE. THE MATHEMATICAL MODEL

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ABSTRACT

The objective of this work was to develop a mathematical model for evaluation of rate of formation of gaseous products of radiolytical and microbial decomposition of cellulose and pressure dependence in fibre reinforced concrete containers (FRCC) contained radioactive wastes at their long-time disposal.

Key words: autradiolysis, biodegradation, radioactive wastes, pressure dependence, time dependence, mathematical models, containers, reinforced concrete, gaseous diffusion, hydrogen, cellulose, radioactive waste disposal

INTRODUCTION

Fibre reinforced concrete containers (FRCC) developed by Sogefibre [1] and produced by VYZKONT Ltd., Trnava are used in Bohunice radioactive waste processing centre (BSC RAO) and deposited at National Radioactive Waste Repository at Mochovce for long-time depository of low-level and intermediate-level radioactive wastes produced in decommissioned A-1 NPP, as well as in Jaslovske Bohunice NPPs and Mochovce NPP. Radioactive wastes immobilised within FRCC contain some amounts of cellulose materials as well as bitumen, metallic wastes and cement grout. These wastes are decomposed by autoradiolysis and also may be decomposed by the microbial decomposing under suitable conditions (presence of microbes and/or moulds as well as humidity).

Physical and physical-chemical parameters of FRCC were published by CHEVALIER [2]. At the long-time depository of radwastes (RW) closed in FRCC the autoradiolysis of cellulose and bitumen wastes proceeds, by which gaseous products are formed.

The review of radiolytical processes of water, cellulose, polymers, bitumen and concrete and microbial degradation of cellulose as well as quantitative model of gas formation in FRCC was prepared [3]. In this work authors concluded that main process for evolving of gaseous products might be microbial degradation of cellulose under suitable conditions, i.e. at higher content of available water, a_w. Under these conditions the pressure may achieve up to 10^5 Pa in dependence of free volume of FRCC.

In the following work [4] we have developed new mathematical model of rate of producing of gaseous products (hydrogen, methane, carbon dioxide) of autoradiolysis and products of microbial decomposition as well as pressure dependence in FRCC. Excess enhance of the pressure inside of FRCC may result in disturbance of FRCC integrity with possible consequential endangering of workers and population health.

In this paper the mathematical model of gas pressure inside of FRCC was educed on the base of physical-chemical parameters of FRCC [1, 2] and of data obtained from VYZ, Slovenske elektrane and UJD SR [3, 4]. Dimensions of FRCC are 1.7 x 1.7 x 1.7 m, with inner dimensions 1.450 x 1.450 x 1.430 m. Thickness of side wall is 0.100 m, bottom 0.125 m and top 0.145 m (mean 0.112 m). Total mass of empty FRCC is 4240 kg. Volumes and amounts of radwastes and other materials, deposited into one FRCC are shown in the tab. 1. The FRCC is filled with cement grout up to 30 mm from upper brink; this represents so-called free volume of FRCC.*

further nutrients no microorganisms were able to grow there during the period of three months. In spite of it in mathematical model we count with extra values of mass portion of biodegradable saccharates in cellulose materials in the range from 13.0% and 100%.

Total radiation-chemical yield of gaseous products formation of radiation decomposition of cellulose for mathematical model was considered conservatively: $G(\text{gas,total})_c$ = 1.9 molecules/100 eV [5]. RYAN [6] presents, that approximate molar ratio of produced gases at radiolytical decomposition of cellulose wastes is $H_2:CO_2 : CO = 1 : 0.7 : 0.3$. In our mathematical model we calculate with ratio of arising gases $H_2 : CO_2 : CO$ = 1:0.42:0.17 and a calculation of cellulose mass decrease is based on balance equation

Tab. 1 The content of one FRCC after filling up [9]

Material	Mass, kg/piece	Number of pieces/FRCC	Total mass, kg
Pressed barrels with biodegradable materials (paper, togs and likewise)	30-50	5	150-200
Barrels with bituminised wastes, 210 kg (20÷40% of radioactive concentrate)	126-168	6 max.	1.020
Cement matrix (~16% mass of salts of radioactive concentrate); ρ=2000 kg.m^{-3}	2.860	1	2.860
Dose rate, $\dot{D}$, Gy s^{-1}	-	-	~5x10^{6}
Free volume under cover of FRCC	~3 cm	-	0.063 dm^3
Free volume in barrels with bitumen, max. 5% (0.2 m^3/barrel)	0.010 m^3	6	0.060 m^3
Number of barrels with available water, a_w(min.)≥0.60	60% barrels	5	P_w – 7.78%

P_w – probability, that in each barrel is $a_w \geq 0,60$

Main gaseous products of microbial anaerobic decomposition of cellulose are CO_2 and methane, what may be expressed by summary equation

$C_6H_{12}O_6 \rightarrow 3CO_2 + 3CH_4$,

thus from one mole of basic unit of cellulose (glucose, 168 g mol^{-1}) arise three moles of CO_2 and three moles of methane. As KOLAROVA [3] found out, in the radwastes containing cellulose, deposited into FRCC in VYZ-SE, mass portion of biodegradable saccharates in cellulose materials (w_{bs}) was in the range from 4.75 to 13.0%. Moreover, under anaerobic conditions without delivery of

$$2C_6H_{12}O_6 = 12H_2 + 5CO_2 + 2CO + 5C$$

i.e. from two moles of decomposed cellulose (basic units, 336 g) arise together 19 moles of gases (0.426 m^3).

Radiation-chemical yield of hydrogen (main gaseous product of bitumen radiation degradation) reaches the value 1.4 molecules/100 eV of absorbed energy [7]. We consider this value in mathematical model of formation of gas and pressure conditions in FRCC. For 2 g-moles of released hydrogen 2 g of total bitumen weight are reduced. At bitumen radiolysis CO, CO_2, CH_4 and hydrocarbon (C_n≥2) are also formed, but rate of their formation is one to two orders lower [8] than rate of hydrogen formation. Therefore in mathematical

* Free volume of FRCC in the present time increases by storage of dilatation layers of extruded polystyrene on the inner side and on bottom of VRCC (2 cm thickness, 5 sides of VRCC).

model we don't consider the mechanism of their formation and influence on weight loss.

Diffusion properties of concrete

A concrete represents porous mass with closed and connected micropores. Concrete permeability and diffusion velocity of gases depend on dimensions and character of joint micropores. The mass transfer through porous materials is called diffusion. It is characterised by diffusion coefficient *D*. It proceeds as a result of thermal motion of molecules. The quantity matter, traversed through surface area unit perpendicular on direction of transfer is called diffusion flow. If in the system exists a concentration gradient *dc/dx* (or *dp/dx*) of some matter in the direction *x,* diffusion flow *I is determined by the first* Ficke law

$$I = -D\frac{dc}{dx}, \tag{3}$$

where *D* is diffusion coefficient, sign minus characterises flow direction from higher to lower concentrations. Value *D* depends on mass and dimension of diffusing particles, surroundings composition and temperature, less on pressure and in the first approach it does not depend on concentration. The rate of matter accumulation at appointed point, conditioned by diffusion, is characterised by the second Ficke law

$$\frac{dc}{dt} = D\frac{d^2c}{dx^2}, \tag{2}$$

where *t* is time. For diffusion in three-dimensional space is in force

$$\frac{dc}{dt} = D\left(\frac{d^2c}{dx^2} + \frac{d^2c}{dy^2} + \frac{d^2c}{dz^2}\right). \tag{3}$$

If in the system the concentration balance (or pressure) of *n* components proceeds, diffusion flow of each component depends on concentration gradient of all other components. In the case of isobaric-isothermal diffusion and also if no external forces are present, then the flow of *i*-component is determined by equation

$$I_i = \sum_{j=1}^{n-1} D_{ij}\frac{\partial c_j}{\partial x} \tag{4}$$

where D_{ij} is diffusion coefficient in a binary mixture of *i*-component with *j*-component(*i* # *j*). Diffusion coefficients of gases in concrete depend on material porosity as well as on gas type.

Radiation decomposition of concrete

Review of radiation-chemical yields of concrete irradiation products was developed in [3]. The main gaseous product is hydrogen. On the base of review [3], the value of radiation-chemical yields of hydrogen was selected for concrete with ^{238}Pu, $G(H_2)=(0.44 \pm 0.03)$ molecules/100 eV [5]. Owing to large mass of cement paste in FRCC (~2860 kg), at higher dose rates the fair-sized volume of gaseous hydrogen can be created, even at lower yields of $G(H_2)$ as selected value. It means, that for specification of calculation it is necessary to know the value $G(H_2)$ in real used cement grout. For 2 g-moles of loosen hydrogen 2 g of total cement grout is reduced.

Dose rate in FRCC

By the modelling of pressure inside of FRCC it is required to give the value of dose rate. On the base of data [3] for cellulose materials 5 μGy s^{-1} was selected as the initial dose rate [9]. Activity of radionuclides immobilized in bitumen and in cement grout provides dose rate around 2 mGy s^{-1} [9]. The assessment of dose rate inside of FRCC and its dependence on deposition time is almost impossible without knowledge of activities of individual radionuclides deposited in FRCC. Thus the ability to analyse the pressure inside of FRCC in dependence on dose rate was included to the mathematical model. In 1999 the FRCC was re-qualified for deposition of activities up to 74 TBq/FRCC [10]. Thus in the mathematical model we consider the dose rate up to 4 mGy s^{-1}, it is possible to set greater extent of dose rates.

It is assumed that the main radionuclide of radwastes is ^{137}Cs with half-life 30.17 year and energy $E_{\beta_{1\max}} = 0.514$ MeV (94.7%), $E_{\beta_{2\max}} = 1.18$ MeV (5.3%) and $E_\gamma = 0.6616$ MeV. The dose can be estimated on the base of method of radioactivity calculation. From the law of radioactive decay results that

$$a_t = a_0 \exp(-\lambda t), \tag{5}$$

where a_0 and a_t are specific activities of radionuclide at the beginning of irradiation and after time expiration t, respectively, and λ is decay constant. If specific activities are given in Bq g^{-1}, $\overline{E}$ is energy of radiation in MeV, t is given in seconds and λ in s^{-1}, then absorbed dose D_t during the time will be equal

$$D_t = 1.602x10^{-10} a_0 \overline{E} \int_0^t \exp(\lambda t) dt \text{, (Gy).} \qquad (6)$$

By integrating it is obtained

$$D_t = \frac{1{,}602.10^{-10} a_0 \overline{E}}{\lambda} \left[1 - \exp(-\lambda t)\right] \text{, (Gy).} \qquad (7)$$

The highest dose is absorbed by system at the moment of complete decay of radionuclide, i.e. at $t \to \infty$ (approximately after 10 half-times)

$$D_\infty = 2.312x10^{-10} a_0 \overline{E} T_{1/2} \text{ (Gy),} \qquad (8)$$

where $T_{1/2}$ is half-time, s.

From the data on radwastes deposited into FRCC [3] results that the total mass of radwastes, m_s, including bitumen and grout reaches: m_c – mass of cellulose in FRCC ≈ 250 kg; m_b + salts – mass of bitumen and radioactive salts ≈ 1020 + 30% salts ≈ 1,326 kg; m_z + salts, mass of cement grout and radioactive salts ≈ 2,860 + 16% salts ≈ 3,318 kg; together m_s ≈ **4,894 kg.**

From the maximal permissible activity 74 TBq/FRCC [10] arises, that specific activity can reach 74 TBq/4,894 kg = 15.12 MBq g^{-1}. By substitution of this specific activity and energy of ^{137}Cs into eq. (6) we obtain the dose rate $\dot{D}_{max}$ ≈ 3.3 mGy s^{-1}, which is the maximal permissible value in FRCC.

EXPERIMENTAL

Starting parameters for mathematical model are given in the Tab. 2 and qualified conservative estimate of critical parameters is in the Tab. 3.

Tab. 2 The initial input parameters

Initial parameter	**Symbol**	**Value**	**Reference**
Diffusion coefficient of j-gas through the walls of FRCC	D_j (j=H_2)	$9.51.10^{-11}$ $m^2.s^{-1}$	[12](Rel.to ^3H)
dtto	D_j (j=CH_4)	$3.36.10^{-11}$ $m^2.s^{-1}$	[12](Rel.to ^3H)
dtto	D_j (j=CO_2)	$2.03.10^{-11}$ $m^2.s^{-1}$	[12](Rel.to ^3H)
dtto	D_j (j=CO)	$2.54.10^{-11}$ $m^2.s^{-1}$	[12](Rel.to ^3H)
Conversion coefficient for radiation-chemical yield	f	$1.04.10^{-7}$ mol.eV.J^{-1}	
Total radiation-chemical yield of gaseous products formation of radiation decomposition of bitumen	G(gas, total)$_b$	1.4 mol-l/100 eV	[7]
Total radiation-chemical yield of gaseous products formation of radiation decomposition of cellulose	G(gas, total)$_c$	1.9 mol-l/100 eV	[5]
Total radiation-chemical yield of gaseous products formation of radiation decomposition of cement grout	G(gas, total)$_z$	0.44 mol-l/100 eV	[5]
Leakage coefficient of FRCC at Δp=0.05 MPa	L	≥0.02 Pa.$m^3.s^{-1}$	[2]
Average thickness of FRCC walls	h	0.112 m	[1]
Initial mass of cellulose waste in FRCC	m_c	250 kg	[9]
Mass of bitumen in FRCC (without salt content)	m_b	1.020 kg	[9]
Mass of grout mixture in FRCC (without salt content)	m_z	2.860 kg	[9]
Initial pressure of gases in free volume of FRCC	p_0	10^5 Pa	
Atmospheric pressure (on surface of FRCC)	p_{atm}	10^5 Pa	
Pressure difference at test of tightness	Δp	$5x10^4$ Pa	[2]
Gas constant	R	8.314 J K^{-1} mol^{-1}	[14]
Inner surface of FRCC	S_{FRCC}	12.5 m^2	
Time increment	t_i	1 year	
Maximal period of deposition of radwastes in FRCC	t_{max}	300 years	[9]
Temperature	T	300 K	[9]
Total inner volume of FRCC	V_o	3.0 m^3	[2]
Molar gas volume	V_A	0.02242 $m^3.mol^{-1}$	[2]
Viscosity of nitrogen (gas)	η	$1.66.10^{-5}$ Pa	[14]
Mass portion of biodegradable saccharates in cellulose materials	w_{bs}	0.13 (max)	[3]

Tab. 3 The qualified conservative assessment of critical parameters

Of material	Parameter	Symbol	Value	Reference
Free volume of FRCC	unfilled free volume of FRCC	V_f	0.0626 m^3	[9]
	free volume in barrels with bitumen (max. 5%), 6 barrels 0,2 m^3	V_b	0.0300 m^3	[9]
	volume of capillary pores in pressurized cellulose (1% of 5x0.0333 m^3)	V_{kc}	0.0017 m^3	
	volume of capillary pores in grout (0.1% of 1.430 m^3)	V_{kz}	0.0014 m^3	
	total free volume in FRCC	V_F	$V_F = V_f + V_c + V_{kc} + V_{kz} \approx 0.0957$ m^3	
Biochemical formation of gases	maximal specific rate of microbial formation of gases	a	$2.3148.10^{-11}$ m^3 kg^{-1} s^{-1}	[15]
Radiation-chemical formation of gases	minimal initial dose rate in FRCC (in cellulose materials)	$\dot{D}_{c(ef)0}$	5.10^{-6} Gy s^{-1}	[3], [9]
	maximal initial dose rate in FRCC (in bitumen)	$\dot{D}_{b(ef)0}$	2.10^{-3} Gy s^{-1}	[9]
	maximal initial dose rate in FRCC (in cement grout)	$\dot{D}_{z(ef)0}$	2.10^{-3} Gy s^{-1}	
	effective half-time of decay (of dominant radionuclide in FRCC)	$T_{1/2(ef)}$	30 years	[9]

Deducing of the mathematical model of gas pressure in the FRCC

On the Fig. 1 the scheme of mathematical model of gas pressure in the FRCC is represented. The maximal specific rate of microbial formation of gases [3] (in anaerobic conditions) is a = 0.002 cm^3 g^{-1} day^{-1}).

The specific rate (mol kg^{-1} s^{-1}) of gaseous products formation of radwastes decomposition in the total volume is

$$\frac{1}{m_s}\frac{dn}{dt} = R = R_m + R_d + R_c \ . \qquad (9)$$

The rate (m^3 s^{-1}) of gaseous products formation of microbial decomposition of biodegradable

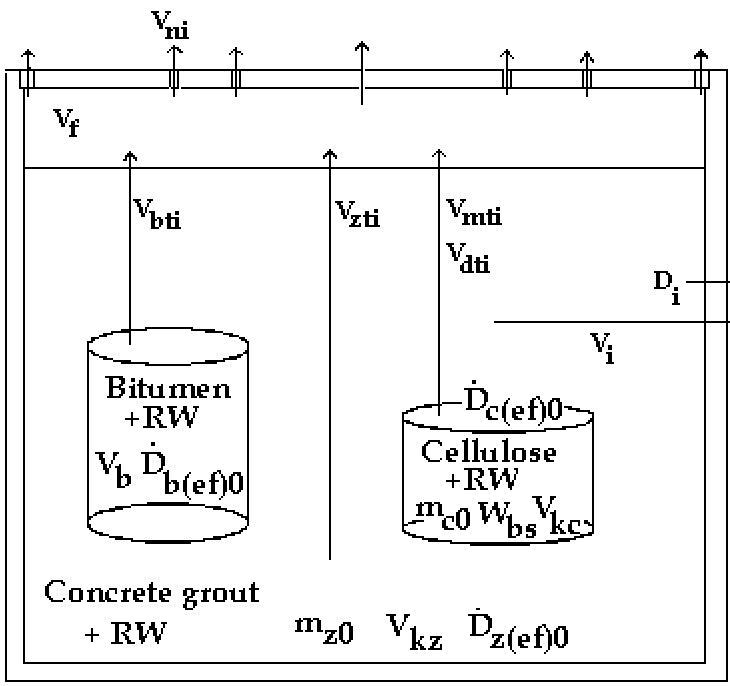

Fig. 1 Scheme of the mathematical model of gas pressure in FRCC (variables are given in Tables 1, 2 and 3)

saccharates of cellulose in FRCC, r_m is

$$r_m = am_c w_{bs} \cdot \quad (10)$$

The volume of gaseous products of microbial decomposition of biodegradable saccharates of cellulose (R_m) arising during the time t_i

$$R_m = V_{mti} = am_c w_{bs} t_i \quad (11)$$

The change of portion of microbial decomposable waste in cellulose after time *i* will be

$$w_{c(t+1)} = \frac{(m_{ct} w_{ct}) - \Delta m_{ct}}{m_{ct}} \quad (12)$$

The rate of radiation decomposition of radwastes depends on half-times decay of radionuclides present in FRCC. Furthermore, as we do not know specific composition of radionuclides and their activities, in the mathematical model we consider effective half-time decay $T_{1/2(ef)}$ and $D_{0(ef)}$ – effective dose at the beginning of irradiation, or $D_{t(ef)}$ – effective dose after time *t*, which can be obtained (it is calculated separately for barrels with cellulose, $D_{c(ef)0}$, barrels with bitumen $D_{b(ef)0,}$ and for cement grout $D_z(ef)_0$) from equation

$$D_{t(ef)} = D_{0(ef)} \exp\left[-\left(\ln 2 / T_{1/2(ef)}\right)t\right] \quad (13)$$

The specific rate of gaseous products formation of radiation decomposition of cellulose r_d in 1 kg of cellulose (mol kg^{-1} s^{-1}) is

$$r_d = fG(gas,total)_c \dot{D}_0 \cdot \quad (14)$$

The rate of gaseous products formation of radiation decomposition of cellulose R_d in FRCC (mol s^{-1}) is

$$R_d = fG(gas,total)_c \dot{D}_0 m_c \quad (15)$$

According to Avogadro law at the temperature 273 K and pressure 0.1 MPa the mole volume of ideal gases is V_A = 0.02242 m^3 mol^{-1}. Thus for *j*-gaseous component equals $V_j = n_j V_A$, therefore volume rate of gaseous products formation of radiation decomposition of cellulose V_d in FRCC (m^3 s^{-1}) is

$$V_d = R_d V_A , \quad (16)$$

V_A is Avogadro number. The volume of created gaseous products of radiation decomposition of cellulose V_{dti} in FRCC during time increment t_i (m^3 r^{-1}) is

$$V_{dti} = R_d V_A t_i \cdot \quad (17)$$

The specific rate of gaseous products formation of radiation decomposition of bitumen r_b in 1 kg of bitumen (mol kg^{-1} s^{-1}) is

$$r_b = fG(gas,total)_b \dot{D}_0 \quad (18)$$

The rate of gaseous products formation (hydrogen is the main gaseous product) of radiation decomposition of bitumen r_d in FRCC (mol s^{-1}) is

$$R_b = fG(gas,total)_b \dot{D}_0 m_b \cdot \quad (19)$$

The volume rate of gaseous products formation of radiation decomposition of bitumen V_b in FRCC (m^3 s^{-1}) is

$$V_b = R_b V_A \cdot \quad (20)$$

The volume of created gaseous products of radiation decomposition of bitumen V_{bti} in FRCC during time increment t_i (m^3 y^{-1}) is

$$V_{bti} = R_b V_A t_i \cdot \quad (21)$$

The specific rate of gaseous products formation of radiation decomposition of cement grout r_z in 1 kg grout (mol kg^{-1} s^{-1}) is

$$r_z = fG(gas,total)_z \dot{D}_0 \cdot \quad (22)$$

The rate of gaseous products formation (hydrogen is the main gaseous product) of radiation decomposition of cement grout R_z in FRCC (mol s^{-1}) is

$$R_z = fG(gas,total)_z \dot{D}_0 m_z \cdot \quad (23)$$

The volume of gaseous products formation of radiation decomposition of cement grout V_z in FRCC (m^3 s^{-1}) is

$$V_z = R_z V_A \cdot \quad (24)$$

The volume of created gaseous products of radiation decomposition of bitumen V_{bti} in FRCC during time increment t_i (m^3 y^{-1}) is

$$V_{zti} = R_z V_A t_i \cdot \quad (25)$$

By assumption, that within the FRCC no chemical reaction of decomposition proceeds, the volume of created gaseous products of chemical decomposition of radwastes, V_{chti} in FRCC during time increment t_i (m^3 y^{-1}) is $V_{chti} = 0$. The total volume of created gaseous products in FRCC during time increment t_i (m^3 y^{-1}) is

$$V_{ti} = V_{mti} + V_{dti} + V_{bti} + V_{zti} + V_{chti} \quad (26)$$

The quantity of decomposed cellulose in FRCC during time increment t_i (from 1 mole of cellulose

$$\Delta m_{cti} = -168 V_{mti} / 6 A_v \ .\qquad (27)$$

The quantity of remaining cellulose (kg) in FRCC after elapse (increment) of time t_i is

$$m_{ci} = m_c + \Delta m_{cti} \ .\qquad (28)$$

The quantity of decomposed bitumen in FRCC during time increment t_i is

$$\Delta m_{bti} = -2 R_b \ .\qquad (29)$$

The quantity of decomposed cement grout in FRCC during time increment t_i is

$$\Delta m_{zti} = -2 R_z \ .\qquad (30)$$

The quantity of moles of gases r_i formed in FRCC during time increment t_i is

$$R_i = R_m + R_d + R_b + R_z \ .\qquad (31)$$

From ideal gas equation

$$pV = nRT \ ,\qquad (32)$$

and at constant free volume V_F inside FRCC

$$V_F = V_f + V_s + V_{kc} + V_k \ ,\qquad (33)$$

and at constant temperature T and at increase of pressure dp_i during a time t_i the rate of pressure increase will be

$$\frac{dp_i}{dt_i} = \frac{RT}{V_F}\frac{dn_i}{dt_i} = \frac{RT}{V_F} R_{ti} \ .\qquad (34)$$

The escape of gases through leakages of the cap of FRCC can be characterized by leakage coefficient. For the FRCC the leaks were tested by the rate of pressure decrease from closed space [2]

$$\frac{dp_i}{dt_i} = \frac{RT}{V_F}\frac{dn_i}{dt_i} = \frac{RT}{V_F} R_{ti} \ .\qquad (35)$$

The value L is necessary to be put out to the used pressure difference Δp = 0.05 MPa and to calculate the permeability of FRCC through leakages

$$\left(dp_n / dt_i \right) = -\left(L / V_0 \right) \ .\qquad (36)$$

Diffusion, by which the concentration (pressure) differences in gas are balanced, depends on mutual collisions among molecules and thus it belongs to transport phenomenon. The substance quantity, which diffuses, according to the 1st Ficke law is proportional to cross-section S, concentration gradient in the diffusion direction –(dc/dx) and dt [11]:

$$dn = -D_j S \left(dc / dx \right) dt \ .\qquad (37)$$

The proportionality constant D_j is the diffusion coefficient of given gas. It depends on mean free line of flight of molecules $\bar{l}$. At diffusion through the porous membranes (if size of pores are small relative to $\bar{l}$) under equal conditions of temperature and pressure, the rate is proportional to the value u_a (u_a – mean rate of molecules A) is in force

$$u_a = \left(2 u_m / \sqrt{\pi} \right) = \sqrt{8RT / \pi M} \ ,\qquad (38)$$

(where u_m – rate of molecules M, M – molecular mass of molecules M), is reciprocal proportional to radix of molecular mass M. The relationship between diffusion rate and motion rate v_A and v_B of two gases A and B through membrane is given by equation

$$\left(D_A / D_B \right) = \left(v_A / v_B \right) = \sqrt{M_B / M_A} \ .\qquad (39)$$

This relation expresses so-called Graham law, according to which the rate of gas diffusion through porous membrane is proportional to radix of its molecular mass. According to [12] the diffusion constant of tritium in concrete is equal to 5.49x10^{-11} m^2 s^{-1}. Relative to molecular mass of tritium and main gaseous products of decomposition H_2, CH_4 and CO_2, according to the relation (39) their diffusion constants in concrete (A = $^3H_2$ = T_2; B = H_2, CH_4, CO_2 and CO) are

$$D_B = D_A \Big/ \sqrt{M_B / M_A} \ ,\qquad (40)$$

respectively: D_{H_2} = 9.51x10^{-11} m^2 s^{-1}, D_{CH_4} = 3.36x10^{-11} m^2 s^{-1}, D_{CO_2} = 2.03x10^{-11} m^2 s^{-1} and D_{CO} = 2.54x10^{-11} $m^2.s^{-1}$ (the values are calculated by us).
The volume rate of diffusion of gases through the walls of FRCC (by assumption, that value D does not depend on pressure difference) will be

$$n_{tij} = v_{tij} = -\left(D_j / h_{VBK} \right) S_{VRCC} t_i \ ,\qquad (41)$$

where j – type of diffusing gas.
The total volume of gas, which diffuses through the walls of FRCC during increment time is

$$V_{Di} = \sum_{j=1}^{j=n} V_{Dij} = -V_A \sum_{j=1}^{j=n} n_{tij} = -V_A \sum_{j=1}^{j=n} n_{tij} \ ,\qquad (42)$$

where j is j-gas. Sign minus means that given volume of gas leaks from FRCC during a year.

From ideal gas equation (32) the decrease of pressure in FRCC can be calculated in consequence of gas diffusion through the walls of FRCC

$$dp_j = n_j \left(RT / V_F \right). \tag{43}$$

The pressure decrease in FRCC in consequence of leakages and diffusion represents during a year

$$dp_t = -\left(dp_n + dp_d \right). \tag{44}$$

If total decrease of pressure in FRCC in consequence of leakages and diffusion is greater than increase of pressure (21), than the pressure in FRCC does not increase.

RESULTS AND CONCLUSIONS

Description of mathematical model of gas pressure in FRCC

Mathematical model of gas pressure in FRCC is programmed in MS Excel 2000™ (the total number of variables is 25). This model was developed in two basic variants:

1. Mathematical model of gas pressure in FRCC as function of dose, Fig. 2.
2. Mathematical model of gas pressure in FRCC as function of mass of cellulose, Fig. 3.

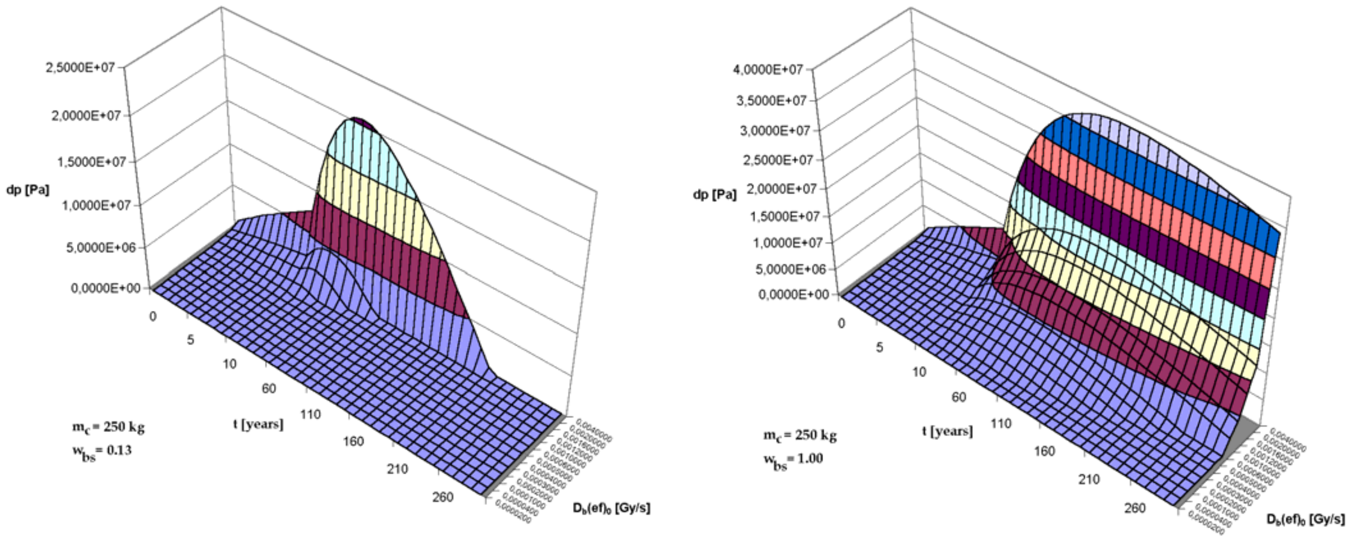

Fig. 2 Pressure simulation inside of FRCC in dependence on dose rate at initial mass of cellulose wastes (m_c) and available water (w_{bs})

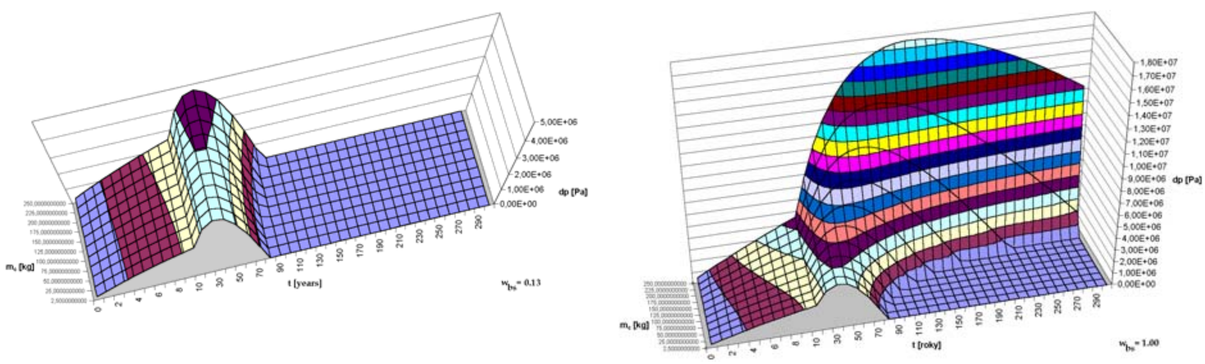

Fig. 3 Pressure simulation inside of FRCC in dependence on initial mass of cellulose m_c and available water (w_{bs})

From the first variant the model ***BITUMEN*** was derived for simulation of gas pressure inside hermetically closed barrel with bituminised radwastes, from the second variant the model ***CELLULOSE*** was derived for simulation of gas pressure inside hermetically closed barrel with cellulose radwastes (for limit case, that surrounding cement grout is gas-tight), Fig. 4.

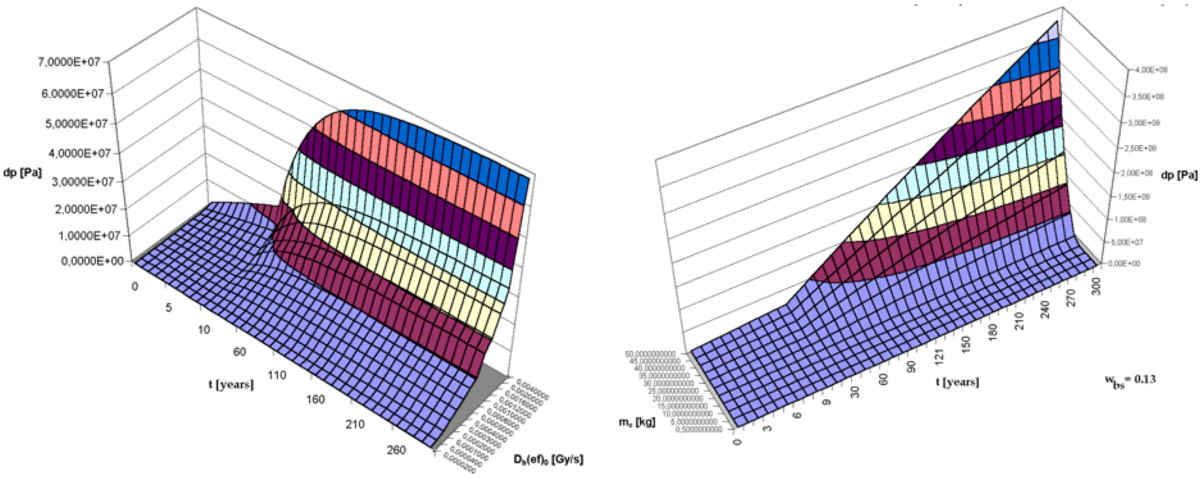

Fig. 4 Pressure simulation inside of barrel with bitumen in dependence on dose rate (SW BITUMEN), and inside of barrel with cellulose wastes in dependence on initial mass of cellulose m_c (with w_{bs} = 0.13, SW CELLULOSE), hermetically sealed in cement grout

- In the first spreadsheet it is possible to input variables into green cells, the values of variables or constants in grey cells cannot be changed.
- The initial minimal dose rate in bitumen can be set, the maximal dose rate in bitumen is calculated by assignment of multiple D(ef)$_0$ by graph creation (12 steps).
- The independent initial minimal dose rate in cement grout can be set, for use of multiplying of dose rate in cement grout by graph creation it is necessary to put the value 1 (in the case $D_{z(ef)0} = D_{b(ef)0}$, then both dose rates are identical in all range of dose rates; in other case only maximal dose rate in cement grout is shown.
- The other variables can be changed after their specifying.

Conclusions

1. The leakages of FRCC are the limiting factor, determining pressure conditions in FRCC. The values of diffusion constants of gases through the walls of FRCC influence the pressure within FRCC relatively slightly.
2. The important factors, which influence the pressure inside FRCC are dose rates in bitumen and in cement grout as well as portion of biologically degradable cellulose.
3. In the case the cement grout is impermeable for gases formed within barrels with cellulose wastes, the pressures could reach up to ~3.2x10^9 Pa; in barrels with bituminised radwastes ~6x10^7 Pa (at dose rate 2 mGy s^{-2}).
4. At the initial mass of cellulose waste in FRCC, $m_{c_{max}}$ = 250 kg, and the maximal mass portion of biodegradable saccharates in cellulose materials, w_{bs} = 0.13 and at $\dot{D} \approx$ 5 μGy s^{-1} in barrels with cellulose wastes and 2 mGy s^{-1} in barrels with bituminised radwastes and in cement grout within FRCC, the maximal pressure could reach approximately ~4.5.10^6 Pa after around 50 years of deposition (Fig. 3).
5. Increase of the initial activity of radionuclides immobilized in barrels with bitumen and in cement grout has the most important influence on the gas formation rate and on pressure conditions inside FRCC. We do not recommend in any case to exceed the initial dose rates over 2 mGy s^{-1}.

6. The experimental measurements have shown [13] that the mastic used for sealing of upper cover and filler gaps in it is not gas-tight and the rate of pressure decrease in FRCC is extensively higher than pressure decrease initiated by diffusion of gases through the walls of FRCC.

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